Electron-hole localization in coupled quantum dots

Filippo Troiani,[1] Ulrich Hohenester,[2, *] and Elisa Molinari[1]

[1] *Istituto Nazionale per la Fisica della Materia (INFM) and Dipartimento di Fisica,*

Università degli Studi di Modena e Reggio Emilia, Via Campi 213/A, 41100 Modena, Italy

[2] *Institut für Theoretische Physik, Karl–Franzens–Universität*

Graz, Universitätsplatz 5, 8010 Graz, Austria

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Abstract

We theoretically investigate correlated electron-hole states in vertically coupled quantum dots. Employing a prototypical double-dot confinement and a configuration-interaction description for the electron-hole states, it is shown that the few-particle ground state undergoes transitions between different quantum states as a function of the interdot distance, resulting in unexpected spatial correlations among carriers and in electron-hole localization. Such transitions provide a direct manifestations of inter- and intradot correlations, which can be directly monitored in experiments.

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Semiconductor quantum dots (QDs) are solid state nanostructures which allow confinement of carriers in all directions within dimensions smaller than their de Broglie wavelength.[1,2] Quantum confinement results in a characteristic discrete energy spectrum and a δ-like density of states; therefore QDs are often referred to as "artificial atoms". Coupling between dots is now becoming an issue of crucial importance. On the one hand, it is an inherent feature of any high-density QD ensemble, as, e.g., needed for most optoelectronic applications.[2] On the other hand, it is essential to the design of (quantum) information devices, for example QD cellular automata[3] or QD implementations of quantum computation.[4] "Artificial molecules" formed by two or more coupled QDs are extremely interesting also from the fundamental point of view, since the interdot coupling can be tuned far out of the regimes accessible in natural molecules, and the relative importance of single-particle tunneling and Coulomb interactions can be varied in a controlled way. The interacting states of N electrons in a double dot were studied theoretically[5,6,7] and experimentally by tunneling and capacitance experiments,[8,9,10,11,12,13] and correlations were found to induce coherence effects and novel ground-state phases depending on the interdot coupling regime.[5,6,7,13]

The effects of correlations on the *photoexcited electron and hole system* in *coupled dots* are instead still largely unknown, in spite of their importance for possible novel applications such as quantum-information processing devices.[4,14] Stacked self-organized dots were demonstrated;[15] exciton splitting in a single artificial molecule was observed in the linear regime, and explained in terms of single-particle level filling of delocalized bonding and anti-bonding electron and hole states.[16,17] When a few photoexcited particles are present, however, Coulomb interactions between electrons and holes adds to the homopolar electron-electron (hole-hole) interactions.[18] The correlated ground and excited states will thus be governed by the competition of these effects, sofar not included in theoretical descriptions of photoexcited artificial molecules. However, even for the simplest symmetric double-dot structures basic questions regarding the nature of electron-hole states are still open. When more than one electron-hole pair is photoexcited, what is the most favorable spatial arrangement of the interacting electrons and holes, i.e., are carriers distributed in both dots or do they prefer localization in the same dot? Can the structural parameters be tuned in order to induce given spatial distributions of electrons and holes in the artificial molecule, thus allowing for a controlled engineering of the Hilbert space? Can electrons and holes tunnel separately or is the system best described in terms of excitonic tunneling?

In this Rapid Communication we analyze ground and excited states of realistic double quantum dots trough a full configuration-interaction description of the few-particle electron-hole system. It is shown that for a given number of excitons the ground state configuration undergoes non-trivial quantum transitions as a function of the interdot distance d resulting in unexpected spatial correlations among carriers, which can be directly monitored in the optical spectra.

The initial ingredients of our calculations are the single-particle wavefunctions $\phi_\mu^m(\boldsymbol{r})$ and energies ϵ_μ^m for electrons ($m = e$) and holes ($m = h$) which are obtained from the solutions of the three-dimensional Schrödinger equation within the envelope-function and effective-mass approximations.[1] We examine a prototypical confinement for two vertically coupled dots, which is double-box-like along z and parabolic in the (x, y)-plane;[19] such parabolic lateral confinement is known to mimic the most important features of various kinds of self-assembled dots and to give results in good agreement with experiment.[1,20,21] The single-particle wavefunctions factorize into in-plane and z-dependent parts, where the in-plane solutions are the well-known Fock-Darwin states.[1] Our approach for the description of the interacting electron-hole states is provided by a full configuration interaction (CI) calculation. Within the Hilbert space of all possible single-particle electron-hole excitations $|\ell\rangle$, the many-body energies E_λ and states $|\lambda\rangle$ are obtained for a given number of electrons and holes by direct diagonalization of the Hamiltonian matrix $\langle\ell|(H_o + H_c)|\ell'\rangle$,[22,23] with H_o the single-particle Hamiltonian and H_c the Coulomb term:

$$H_c = \frac{1}{2} \sum_{mn} \int d(\boldsymbol{r}\boldsymbol{r}') \frac{\psi_m^\dagger(\boldsymbol{r})\psi_n^\dagger(\boldsymbol{r}')\psi_n(\boldsymbol{r}')\psi_m(\boldsymbol{r})}{\kappa|\boldsymbol{r} - \boldsymbol{r}'|}, \tag{1}$$

which accounts for all possible electron-electron, hole-hole, and electron-hole interactions; here the field operators $\psi_m^\dagger(\boldsymbol{r})$ create an electron or hole at position $\boldsymbol{r}$, and κ is the semi-conductor dielectric constant. In our calculations truncation of the Hilbert space to the 12 single-particle states of lowest energy for both electrons and holes is found to yield very accurate convergence for the lowest few-particle states.

Let us first analyze the single-particle (SP) ingredients of our coupled-QD model. Figure 1 shows the energies for the "bonding" (solid line) and "antibonding" (dashed line) state of lowest energy for electrons (left axis) and holes (right axis), respectively; the insets show the wavefunctions along z for two selected values of the interdot distance d. Because of the

heavier hole mass hole tunneling becomes suppressed at smaller interdot distances d than electron tunneling; this is also reflected in the faster decrease of the bonding-antibonding splitting with increasing d.

If we would neglect at the lowest order of approximation Coulomb interactions, the few-particle electron-hole states would be simply obtained by filling successive SP electron and hole states. However, inclusion of Coulomb interactions gives rise to a mixing of different SP states, where the gain in potential energy through Coulomb correlations is achieved at the price of populating states with higher SP energies. The few-particle electron-hole states thus result from the detailed interplay of these trends, where the relative importance of SP energies and the increase of the Coulomb ones varies with interdot distance d. As will be shown in the following, such interplay can drastically alter the simple-minded SP picture.

We first consider the case of a single electron-hole pair (exciton). The diamonds in Fig. 2 show the d-dependence of the exciton groundstate. An estimate of Coulomb correlations is provided by the usual electron-hole correlation function

$$g_{eh}(\boldsymbol{r}_e, \boldsymbol{r}_h) = \langle \psi_e^\dagger(\boldsymbol{r}_e)\psi_h^\dagger(\boldsymbol{r}_h)\psi_h(\boldsymbol{r}_h)\psi_e(\boldsymbol{r}_e)\rangle, \tag{2}$$

which gives the probability of finding an electron at position $\boldsymbol{r}_e$ when a hole is at position $\boldsymbol{r}_h$. In the insets of Fig. 2 we plot the spatial average of g_{eh} over the center-of-mass coordinate $\boldsymbol{R} = \frac{1}{2}(\boldsymbol{r}_e + \boldsymbol{r}_h)$, $\bar{g}_{eh}(\boldsymbol{r})$, which instead gives the probability of finding the two particles at the relative position $\boldsymbol{r} = \boldsymbol{r}_e - \boldsymbol{r}_h$. Owing to the strong Coulomb interaction the electron and hole stay together and $\bar{g}_{eh}$ is localized around $\boldsymbol{r} \approx \boldsymbol{0}$ [insets in Fig. 2, with crosses at $\boldsymbol{r} = \boldsymbol{0}$]. Thus, *even for the smallest interdot distance d exciton tunneling dominates over separate electron and hole tunneling.* Note that in a pure single-particle picture, with electrons and holes occupying the lowest available SP states, there would be no spatial correlation between them; this would be reflected in $\bar{g}_{eh}(x, 0, z)$ by a structure with two peaks around $z = \pm d$ and a larger one centred on $z = 0$ (for an electron in the left dot the hole is with equal probability in the left or right dot, and vice-versa).

The effects of Coulomb-correlations become more striking when higher-order interactions —i.e. beyond single excitons— are taken into account. In contrast to transport experiments,[8,9,10,11,12,13] which primarily give information about groundstate properties, optical spectroscopy allows easy access to excited states. For instance, in non-linear coher-

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ent spectroscopy[25] a strong "pump" laser prepares the system in an exciton state, and a weak "probe" beam measures the optical transitions to biexciton states (i.e., two Coulomb-correlated electron-hole pairs). The solid lines in Fig. 2 are associated with the transitions from the exciton groundstate to the lowest optically active biexciton states where the two electron-hole pairs have parallel (gray line) or opposite (black lines) spin orientations; the thickness of the lines is proportional to the oscillator strengths of the respective transitions.[22] As to the general trends of the curves, the most striking features are the following: for parallel spins, because of Pauli blocking the second electron-hole pair is created in an excited state, as reflected by the increase of the optical transition energy with increasing d ("antibonding" behaviour). On the contrary, for antiparallel spins the overall d-dependence of the lowest biexciton states shows a "bonding" behavior, which reflects the predominant occupation of the SP "bonding" states. More in detail, one can see that at a critical inter-dot distance $d_c \approx 2$ nm an anti-crossing occurs between the two solid black lines, where the energetically lowest one picks up all the oscillator strength. Besides, at the largest interdot distances, $d \cong 3$ nm, two of the lines merge with the exciton energy: the corresponding biexciton states consist of two spatially separated excitons localized in the two dots, as discussed below. At the same values of d, in the biexciton groundstate all four particles are localized in the same dot, where because of Coulomb correlations the energy is reduced by ≈ 1 meV as compared to the uncorrelated case (biexciton binding energy).[20,22]

In the following we analyze in more detail the lowest biexcitonic states. As will be apparent from the results of Fig. 3, there exist two major trends for correlation effects: In case of "anti-correlation" electrons and holes tend to avoid each other, and the repulsion energy is minimized when each dot is populated by one electron-hole pair. As in the absence of external fields there is no net charge distribution in each QD, there exists an only very weak long-range interaction between the excitons—even in regimes where the QDs are tunneling-coupled (Fig. 1); as consequence, the biexcitonic energy at large d tends to twice the excitonic one (grey and upper black curve, respectively, in Fig. 2). Alternatively, correlation can favour spatial arrangements where all four particles are localized in the same QD. This requires a correlation of higher order[24] with occupation of the antibonding states for both electrons and holes. When all particles are localized in the same QD, in-plane spatial correlations becomes effective which give rise to a short-range interaction (analogue to a classical interaction between induced dipoles) and to the well-known biexcitonic binding

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energy ΔE_{2X}. This spatial arrangement becomes energetically more favourable with respect to the former when the cost of its stronger degree of correlation in terms of SP energy is $\lesssim \Delta E_{2X}$.

Figure 3 shows the d-dependence of the (a) energies, and the mean (b) electron-hole, (c) electron-electron, and (d) hole-hole distances for the four biexciton states of lowest energy. Let us first focus on the ground state. At the smallest values of d, the ground state is seen to correpond to: no electron-hole correlation, intermediate anti-correlation between electrons and a pronounced one between holes [left insets in Figs. 3 (b-d)]. This different degree of correlation for electrons and holes can be attributed to the different SP level splittings (Fig. 1) and the corresponding different cost in SP energy to create two-particle correlations. Apparently, at d_c the system undergoes an abrupt transition. The correlation functions [middle and right insets of Figs. 3 (b-d)] show how spatial correlation becomes more and more effective, with strong correlations between all particles. Such transition is also confirmed by the trends of the mean carrier-carrier distances that abruptly decrease for $d \approx d_c$. The second optically active biexcitonic state (continuous grey lines) shows a precisely reversed trend (and a similar anti-crossing occurs between the two intermediate, non optically active states [dotted lines in Figs. 3(a-d)].

Further calculations show that the distance at which the transition occurs can be tailored by controlling the QD confinement and interdot coupling. In addition, we have found that the effects are preserved in the presence of small asymmetries between both dots, although the transitions occur at different distance d for different states, and that similar phenomena persist for a larger number of photoexcited particles in the double dot.[26].

In conclusion, we have addressed for the first time few-particle correlations in photoexcited coupled quantum dots. We have demonstrated that they can induce novel transitions between different quantum states as a function of the interdot distance d, and are essential for a quantitative or even qualitative description of the electron and hole states. The fundamental reason for these transitions lies in a very general phenomenon, namely that interdot coupling controls the balance between the gain in Coulomb energy obtained by the spatial correlation of carriers (which requires occupation of higher-lying single-particle states) and the corresponding cost in single-particle energy. These transitions leave a clear fingerprint in the nonlinear optical spectra. Beside their fundamental interest as manifestations of inter- and intradot correlations, these quantum transitions may be used to tailor the Hilbert space

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structure and the qubit identification in quantum computation schemes based on coherent optical control of coupled quantum dots.

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[*] Electronic address: ulrich.hohenester@uni-graz.at

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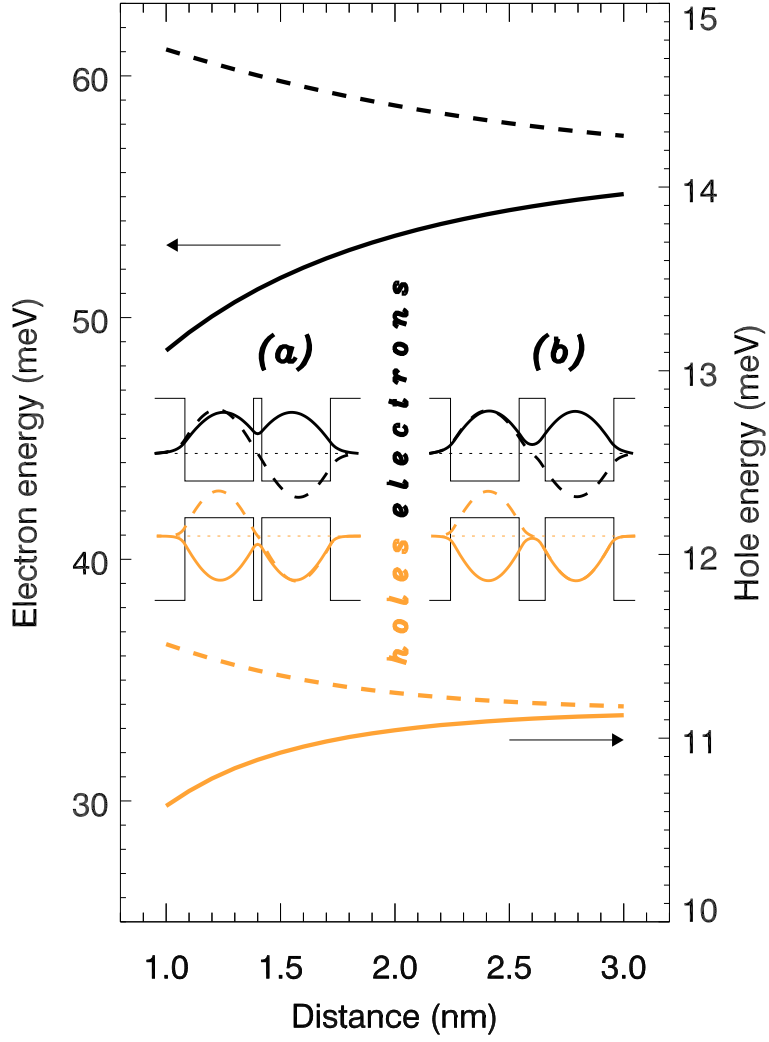[26] F. Troiani, U. Hohenester and E. Molinari, to be published.

FIG. 1: Single-particle energies for electrons (left axis) and holes (right axis) in a symmetric double QD as a function of the interdot distance d. The curves correspond to the lowest Fock-Darwin state and to the bonding (solid line) and antibonding (dashed line) groundstates along z. The insets show the electron and hole wavefunctions along z for two selected interdot distances of 1.2 nm [(a)] and 2.8 nm [(b)], respectively.
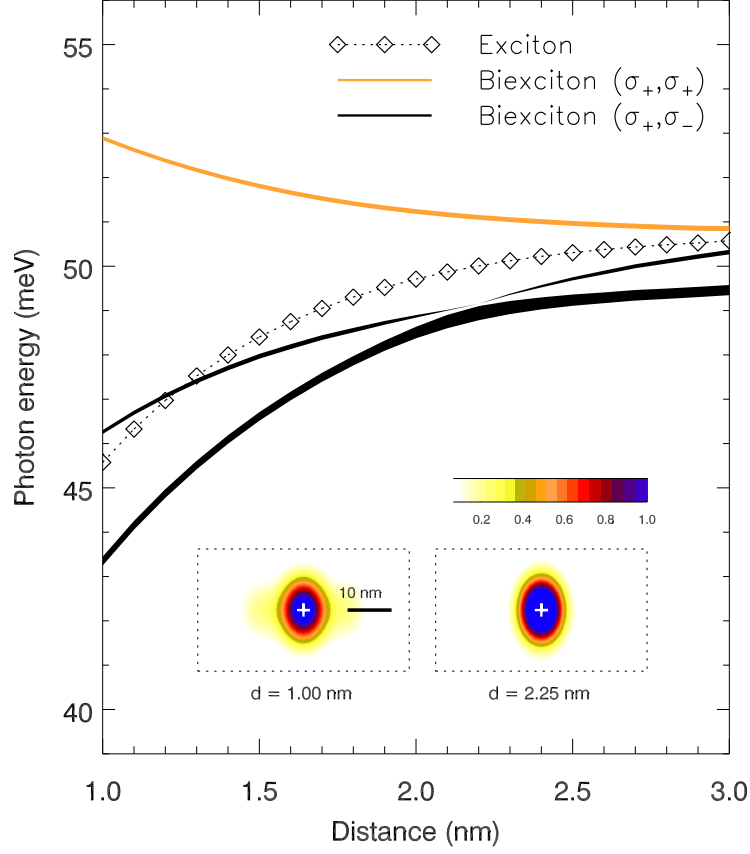
FIG. 2: Excitation energies of a symmetric double QD. Diamonds: Exciton energies as a function of interdot distance d. Solid lines: Transition energies from the exciton groundstate to the biexciton state where the two electron-hole pairs have parallel (gray line) or antiparallel spins (black lines); the thickness of the lines corresponds to the oscillator strengths of the corresponding transitions. Insets: Normalized electron-hole correlation function $\bar{g}_{eh}(x, 0, z)$ (x and z in vertical and horizontal directions, respectively) for the exciton groundstate; the crosses indicate $x = z = 0$.
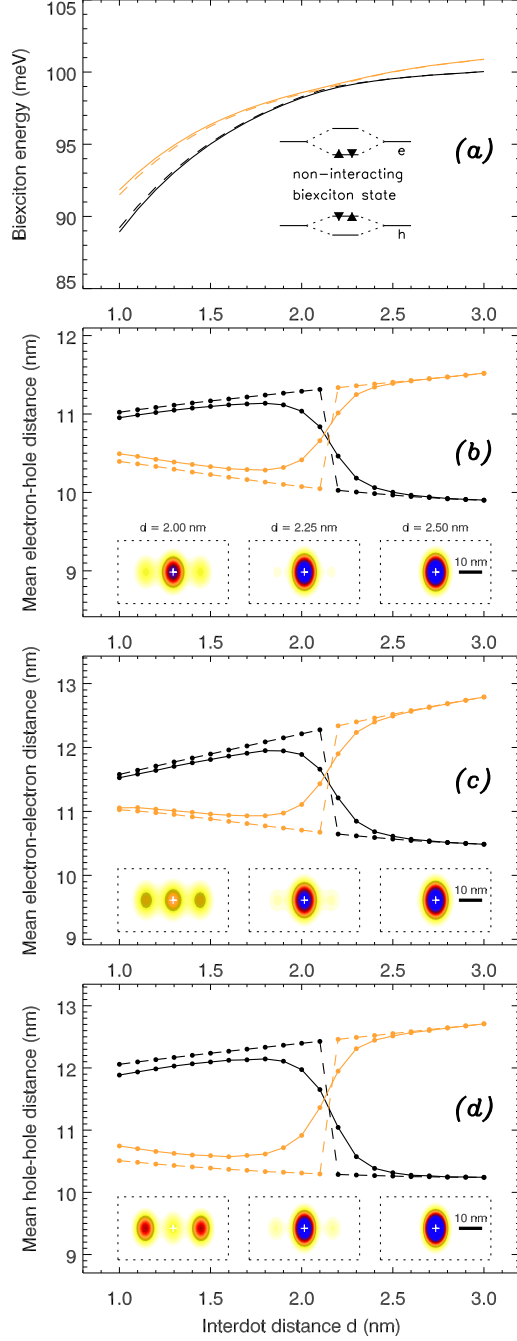
FIG. 3: Biexciton energies [(a)] and mean distances between particles [(b–d)] in a symmetric double QD. The four lowest energetic levels are considered. The continuos curves refer to the optically active biexciton states involved in the transitions of Fig. 2 (black lines). Insets: Electron-hole [(b)], electron-electron [(c)] and hole-hole [(d)] correlation functions $\bar{g}(x, 0, z)$.

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